

# The characterization of high-pressure superconducting state in $\text{Si}_2\text{H}_6$ compound: the strong-coupling description

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The thermodynamic parameters of the superconducting state, that induces in  $\text{Si}_2\text{H}_6$  under the pressure at 275 GPa, have been calculated. In the framework of the Eliashberg formalism, it has been shown that the critical temperature can attain extremely high values:  $T_C (\mu^*) \in \langle 173.36, 99.99 \rangle$  K, where the Coulomb pseudopotential ( $\mu^*$ ) belongs to the range from 0.1 to 0.3. The ratio of the energy gap to the critical temperature ( $R_\Delta$ ) significantly exceeds the value predicted by the BCS theory:  $R_\Delta (\mu^*) \in \langle 4.40, 4.04 \rangle$ . Additionally, it has been stated that in the whole range of the superconducting state's existence, the electron effective mass ( $m_e^*$ ) is large;  $[m_e^*]_{T=T_C}^{\max} = 2.397m_e$ , where the symbol  $m_e$  denotes the electron band mass.

Keywords:  $\text{Si}_2\text{H}_6$ -superconductor, High-pressure effects, Thermodynamic properties.

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The induction of the superconducting state with the high value of the critical temperature ( $T_C$ ) is one of the fundamental problems of the modern solid state physics.

The conducted investigations allowed to determine that the electron-phonon interaction in the two-band system could lead to the formation of the superconducting phase with the relatively high critical temperature; the maximum of  $T_C$  has been discovered for  $\text{MgB}_2$  (under normal conditions), where the critical temperature is equal to 39.4 K [1]. It should be noticed that the remaining thermodynamic parameters of the superconducting state differ significantly from the predictions of the BCS theory [2], [3], [4], [5].

As for now, the highest values of the critical temperature can be observed in the cuprates discovered in 1986 by Bednorz and Muller [6]. In particular, for the compound  $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$  under the normal conditions,  $T_C = 135$  K has been stated [7]. Under the influence of the pressure  $p \simeq 31$  GPa, the critical temperature in  $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$  increases up to the value of about 164 K [8]. Due to the lack of the generally acceptable theory of the superconducting state in the cuprates, it is difficult to say how high value may take  $T_C$ . However, some clues can be provided by the recently achieved results which are presented in the paper [9].

Another direction of the studies is being connected with the possibility of the induction of the high-temperature superconducting state in the classical systems that stay under the influence of the high pressure. The strong argument for this type of research are the experimental results obtained for lithium and calcium. In the case of lithium, it has been found  $[T_C]_{\max} = 14$  K for  $p = 30.2$  GPa, whereas for calcium  $[T_C]_{\max}$  equals 25 K ( $p = 161$  GPa) [10], [11]. However, Sakata *et al.* have suggested that the value of the critical temperature for calcium can reach 29 K ( $p = 216$  GPa) [12] - but this result has been challenged by Andersson [13]. A full review of an anomalous thermodynamic properties of the superconducting state in lithium, calcium and  $\text{CaLi}_2$  can be found by the readers in papers: [14], [15], [16], [17], [18], [19].

From the theoretical point of view, the highest value of the critical temperature should characterize the superconducting state in the metallic hydrogen [20]. The numerical calculations performed in the pressure range from  $\sim 400$  GPa to 3.5 TPa revealed that  $T_C$  is extremely high [21], [22], [23], [24], [25]. In particular, for  $p = 2$  TPa the critical temperature changes from 413 K to 631 K (depending on the assumed Coulomb pseudopotential) [26].

The above theoretical results, although very interesting, are now experimentally unverifiable because of the high pressure metallization ( $p_m \sim 400$  GPa) [27]. For this reason, the way for the reduction of  $p_m$  is being sought out. A good suggestion seems to be the use of the chemical pre-compression [28]. The recently conducted studies allowed to figure out that the metallization pressure for  $\text{SiH}_4$  is about 55 GPa [29], [30]. What is more important - the compound  $\text{SiH}_4$  goes into the superconducting state with the critical temperature of 17 K for the pressure 96 GPa and 120 GPa [30]. The theoretical calculations suggest even higher values of the critical temperature for the system  $\text{Si}_2\text{H}_6$  crystallizing in the structure  $Pm - 3m$  at 275 GPa [31] and for the compound  $\text{SiH}_4(\text{H}_2)_2$  - the structure  $Ccca$  at 250 GPa [32].

In the paper, the thermodynamic parameters of the superconducting state in  $\text{Si}_2\text{H}_6$  ( $p = 275$  GPa) have been determined. Due to the high value of the electron-phonon coupling constant ( $\lambda = 1.4$ ), the calculations have been

carried out in the framework of the strong-coupling formalism (the Eliashberg approach) [33], [34], [35], [36].

The Eliashberg equations in the mixed representation have the following form [37]:

$$\begin{aligned} \phi(\omega + i\delta) = & \frac{\pi}{\beta} \sum_{m=-M}^M [\lambda(\omega - i\omega_m) - \mu^* \theta(\omega_c - |\omega_m|)] \frac{\phi_m}{\sqrt{\omega_m^2 Z_m^2 + \phi_m^2}} \\ & + i\pi \int_0^{+\infty} d\omega' \alpha^2 F(\omega') \left[ [N(\omega') + f(\omega' - \omega)] \frac{\phi(\omega - \omega' + i\delta)}{\sqrt{(\omega - \omega')^2 Z^2(\omega - \omega' + i\delta) - \phi^2(\omega - \omega' + i\delta)}} \right] \\ & + i\pi \int_0^{+\infty} d\omega' \alpha^2 F(\omega') \left[ [N(\omega') + f(\omega' + \omega)] \frac{\phi(\omega + \omega' + i\delta)}{\sqrt{(\omega + \omega')^2 Z^2(\omega + \omega' + i\delta) - \phi^2(\omega + \omega' + i\delta)}} \right], \end{aligned} \quad (1)$$

and

$$\begin{aligned} Z(\omega + i\delta) = & 1 + \frac{i\pi}{\omega\beta} \sum_{m=-M}^M \lambda(\omega - i\omega_m) \frac{\omega_m Z_m}{\sqrt{\omega_m^2 Z_m^2 + \phi_m^2}} \\ & + \frac{i\pi}{\omega} \int_0^{+\infty} d\omega' \alpha^2 F(\omega') \left[ [N(\omega') + f(\omega' - \omega)] \frac{(\omega - \omega') Z(\omega - \omega' + i\delta)}{\sqrt{(\omega - \omega')^2 Z^2(\omega - \omega' + i\delta) - \phi^2(\omega - \omega' + i\delta)}} \right] \\ & + \frac{i\pi}{\omega} \int_0^{+\infty} d\omega' \alpha^2 F(\omega') \left[ [N(\omega') + f(\omega' + \omega)] \frac{(\omega + \omega') Z(\omega + \omega' + i\delta)}{\sqrt{(\omega + \omega')^2 Z^2(\omega + \omega' + i\delta) - \phi^2(\omega + \omega' + i\delta)}} \right]. \end{aligned} \quad (2)$$

The symbols  $\phi(\omega)$ ,  $Z(\omega)$  and  $(\phi_m \equiv \phi(i\omega_m)$ ,  $Z_m \equiv Z(i\omega_m))$  denote the order parameter function and the wave function renormalization factor on the real (imaginary) axis, respectively;  $\omega_m$  represents the  $m$ -th Matsubara frequency:  $\omega_m \equiv (\pi/\beta)(2m-1)$ , where  $\beta \equiv (k_B T)^{-1}$  ( $k_B$  is the Boltzmann constant). The order parameter is defined with an expression:  $\Delta \equiv \phi/Z$ .

The functions  $\phi_m$  and  $Z_m$  should be calculated by solving the Eliashberg equations on the imaginary axis:

$$\phi_m = \frac{\pi}{\beta} \sum_{n=-M}^M \frac{\lambda(i\omega_m - i\omega_n) - \mu^* \theta(\omega_c - |\omega_n|)}{\sqrt{\omega_n^2 Z_n^2 + \phi_n^2}} \phi_n, \quad (3)$$

$$Z_m = 1 + \frac{1}{\omega_m} \frac{\pi}{\beta} \sum_{n=-M}^M \frac{\lambda(i\omega_m - i\omega_n)}{\sqrt{\omega_n^2 Z_n^2 + \phi_n^2}} \omega_n Z_n. \quad (4)$$

The pairing kernel for the electron-phonon interaction is given by:  $\lambda(z) \equiv 2 \int_0^{\Omega_{\max}} d\Omega \frac{\Omega}{\Omega^2 - z^2} \alpha^2 F(\Omega)$ , where  $\alpha^2 F(\Omega)$  means the Eliashberg function. For  $\text{Si}_2\text{H}_6$ , the Eliashberg function has been calculated in [31]; the maximum phonon frequency ( $\Omega_{\max}$ ) is equal to 284.41 meV.

The Coulomb interaction, occurring between the electrons, is parametrized using the Coulomb pseudopotential  $\mu^*$ . The symbol  $\theta$  denotes the Heaviside function;  $\omega_c$  is the cut-off frequency ( $\omega_c = 3\Omega_{\max}$ ).

The symbol  $N(\omega)$  and  $f(\omega)$  represents the Bose-Einstein and Fermi-Dirac function, respectively. The Eliashberg equations have been solved with the help of the numerical methods used in the papers: [38], [39], [40], [41]. The convergence of the solutions has been obtained for  $T \geq T_0 = 23.21$  K ( $M = 1100$ ).

The solutions of the Eliashberg equations have been analyzed for  $\mu^* \in \langle 0.1, 0.3 \rangle$ . The results obtained for the imaginary axis have been presented in Fig. 1. In particular, in the first row there is plotted a dependence of the order parameter on the temperature and the Coulomb pseudopotential. The second row presents the results for the wave function renormalization factor.

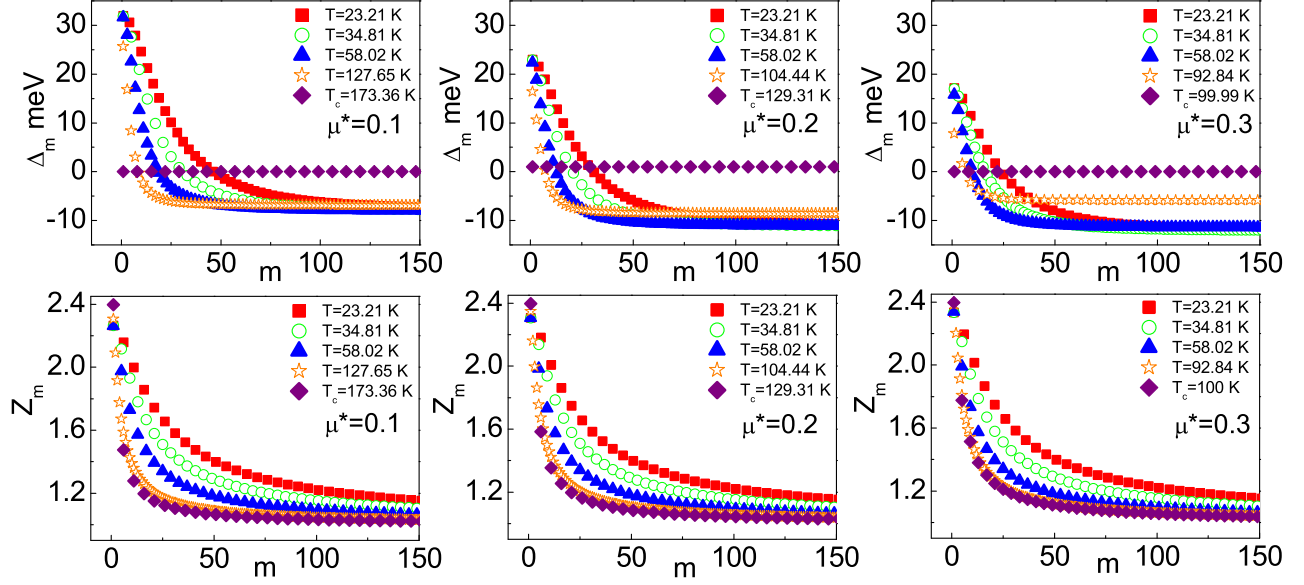


FIG. 1: The solutions of the Eliashberg equations on the imaginary axis for the selected values of the temperature and the Coulomb pseudopotential. The first 150 values of  $\Delta_m$  and  $Z_m$  have been shown.

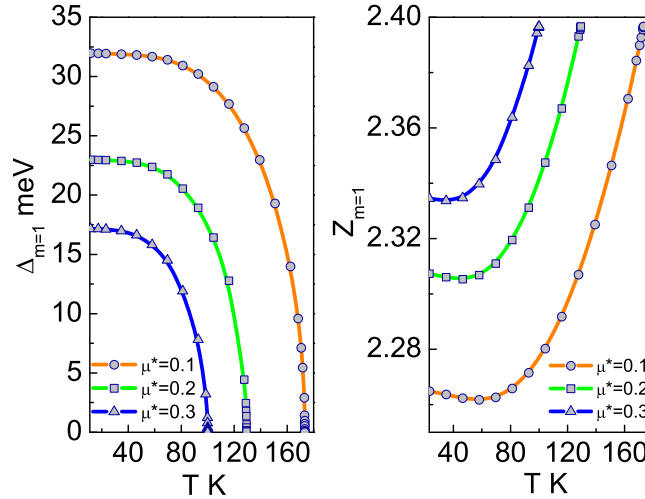


FIG. 2: The dependence of  $\Delta_{m=1}$  and  $Z_{m=1}$  on the temperature for the selected values of the Coulomb pseudopotential.

On the basis of Fig. 1, it is easy to notice that the values of the function  $\Delta_m$  strongly decrease together with the increase of  $m$ . The increase of the temperature and the Coulomb pseudopotential causes also a decrease in the value of the parameter  $\Delta_m$ .

In the case of the wave function renormalization factor, the growth of the number  $m$  results in the decrease in the value of  $Z_m$ . However, the wave function renormalization factor very weakly depends on the temperature and the Coulomb pseudopotential.

The full dependence of the order parameter and the wave function renormalization factor on the temperature for the selected values of the Coulomb pseudopotential can be most conveniently traced after plotting the shape of the function  $\Delta_{m=1}$  and  $Z_{m=1}$  (see Fig. 2). In particular, the maximum value of the order parameter can be parametrized with the help of the following expression:  $\Delta_{m=1}(T, \mu^*) = \Delta_{m=1}(T_0, \mu^*) \sqrt{1 - \left(\frac{T}{T_0}\right)^\beta}$ , where  $\Delta_{m=1}(T_0, \mu^*) = 157.54(\mu^*)^2 - 137.11\mu^* + 44.11$  meV and  $\beta = 3.4$ .

In the next step, the dependence of  $T_C$  on  $\mu^*$  has been precisely determined. On the basis of the results presented in Fig. 3, it is easy to notice that the critical temperature is very high in the whole range of the considered

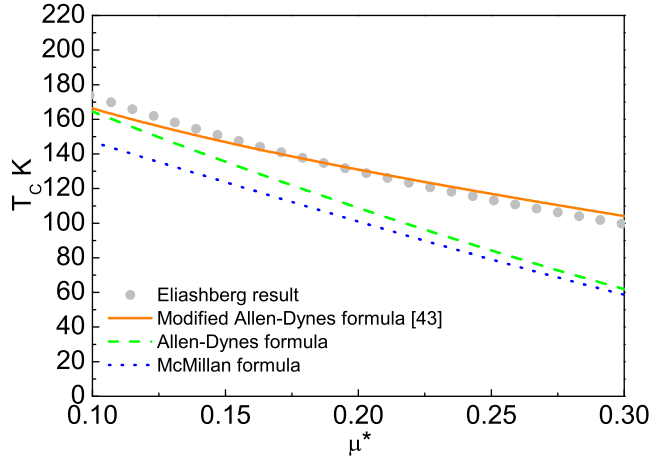


FIG. 3: The dependence of the critical temperature on the values of the Coulomb pseudopotential, determined with an use of the selected methods.

values of the Coulomb pseudopotential;  $T_C \in \langle 173.36, 99.99 \rangle$  K. Let us note that the calculated critical temperatures ( $T_C(\mu^* = 0.1) = 173.36$  K and  $T_C(\mu^* = 0.13) = 158.57$  K) significantly exceed the values determined in the paper [31]:  $T_C(\mu^* = 0.1) = 153.44$  K and  $T_C(\mu^* = 0.13) = 138.86$  K. The obtained result is related to the fact that  $T_C$  in [31] has been estimated on the basis of the classical Allen-Dynes formula, which significantly lowers its value.

Generalizing the achieved result, Fig. 3 presents the complete dependence of the critical temperature on the Coulomb pseudopotential calculated on the basis of the classical expressions of Allen-Dynes and McMillan [42], [43]. It can be clearly seen that with the increase of  $\mu^*$  the critical temperature, based on classical formulas, more visibly differs from the strict result based on the Eliashberg equations.

Let us notice that the function  $T_C(\mu^*)$  can be determined with a good approximation using the analytical formula. However, it is necessary to use the formula originally derived for  $\text{SiH}_4(\text{H}_2)_2$  [44].

The Eliashberg equations in the mixed representation have been solved for the identical range of the temperatures and the Coulomb pseudopotential as the Eliashberg equations on the imaginary axis.

Fig. 4 presents the form of the order parameter on the real axis in the frequency range from 0 to  $\Omega_{\max}$ . Additionally, the rescaled Eliashberg function ( $30\alpha^2F(\Omega)$ ) has been also plotted. It can be easily seen that for low temperatures, the course of the order parameter is far more complex than for the higher temperatures. The difference stems from the fact that in the range of the low temperatures the form of the function  $\Delta(\omega)$  is strongly correlated with the complicated shape of the Eliashberg function [45].

It is also convenient to plot the order parameter on the complex plane. In particular, Fig. 5 presents the values of  $\Delta(\omega)$  in the dependence on the temperature and the Coulomb pseudopotential. The wide range of the frequencies has been selected:  $\omega \in \langle 0, \omega_c \rangle$ . It can be noticed that the values of the order parameter lay down on the characteristic spirals with the radius decreasing together with the increase of  $T$  and  $\mu^*$ . Note that similar spirals for the order parameter have been observed for Pb, Hg and Sn in the paper [46].

The courses plotted in Fig. 5 allow to determine the frequency range for which the effective potential of the electron-electron interaction is pairing. From the mathematical point of view, the values of the above frequencies are calculated on the basis of the following condition:  $\text{Re}[\Delta(\omega)] > 0$  [45]. For  $\mu^* = 0.1$ , the range of the frequency corresponding to the pairing potential extends from 0 to  $\omega_p \simeq 1.2\Omega_{\max}$ . With the increase of the Coulomb pseudopotential  $\omega_p$  decreases. However, even for the high value of the pseudopotential ( $\mu^* = 0.3$ ) occurs  $\omega_p > \Omega_{\max}$ . The above result is related to the fact that the electron-phonon coupling constant for  $\text{Si}_2\text{H}_6$  takes the high value.

Basing on the presented results, the low-temperature value of the energy gap  $2\Delta(0)$  at the Fermi level ( $T = T_0$ ) has been calculated. In particular, the following expression has been used:  $\Delta(T) = \text{Re}[\Delta(\omega = \Delta(T))]$ . As a result it has been obtained:  $2\Delta(0) \in \langle 65.73, 34.85 \rangle$  meV for  $\mu^* \in \langle 0.1, 0.3 \rangle$ .

The knowledge of the energy gap's value allows to determine the dimensionless ratio  $R_\Delta \equiv 2\Delta(0)/k_B T_C$ . For compound  $\text{Si}_2\text{H}_6$  it has been achieved:  $R_\Delta \in \langle 4.40, 4.04 \rangle$ . The above result is really far from the  $R_\Delta$  predicted by the BCS theory;  $[R_\Delta]_{\text{BCS}} = 3.53$  [4], [5]. Let us notice that the dependence of  $R_\Delta$  on  $\mu^*$  can be determined with a good accuracy based on an analytical formula derived for the compound  $\text{SiH}_4(\text{H}_2)_2$  [47].

Second solution of the Eliashberg equations allows to determine the electron effective mass:  $m_e^* = \text{Re}[Z(\omega = 0)] m_e$ , where symbol  $m_e$  stands for the band mass. After the appropriate calculations, it has been stated that the quantity

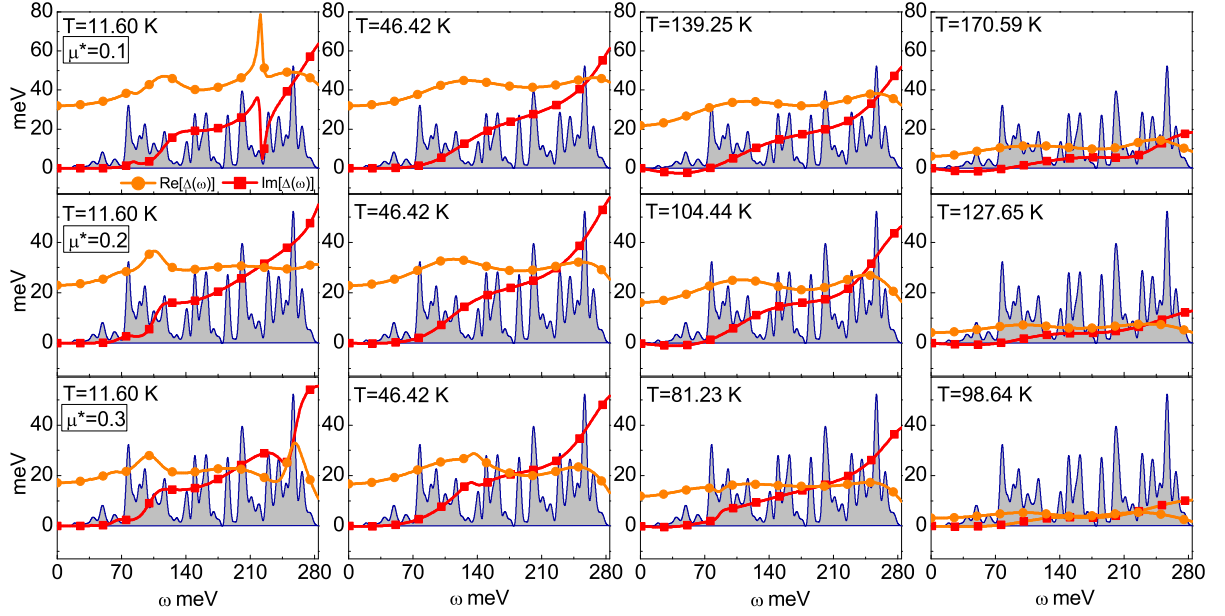


FIG. 4: The real and imaginary part of the order parameter on the real axis for the selected values of the temperature and the Coulomb pseudopotential. Additionally, the rescaled Eliashberg function has been plotted.

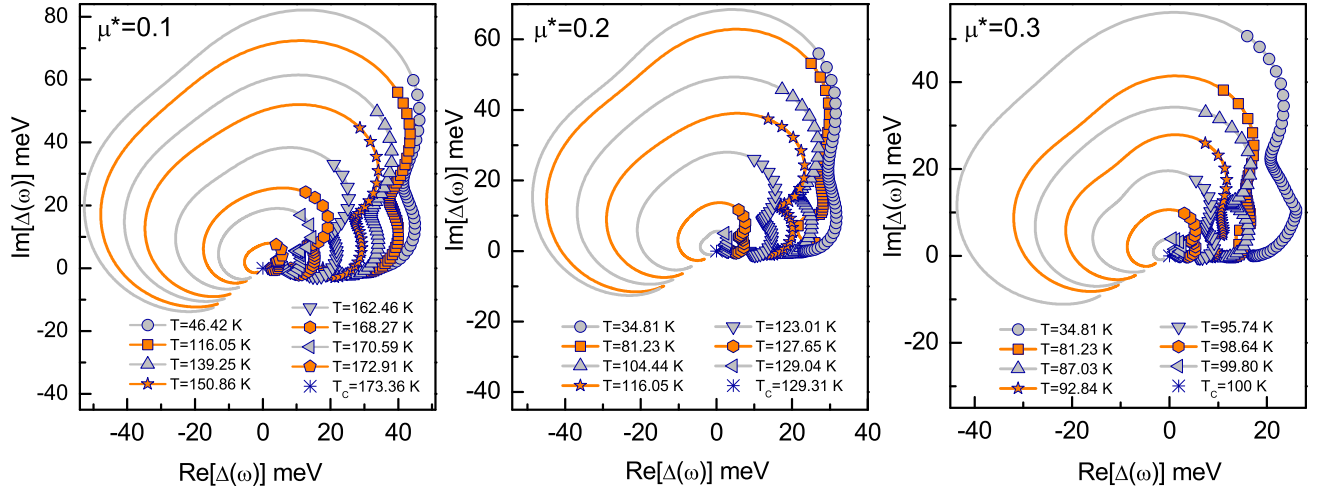


FIG. 5: The order parameter on the complex plane for the selected values of the temperature and the Coulomb pseudopotential. The lines with symbols have been achieved for  $\omega \in (0, \Omega_{\max})$ ; the lines without symbols correspond to the frequencies' range from  $\Omega_{\max}$  to  $\omega_c$ .

$m_e^*$  weakly depends on the temperature and the Coulomb pseudopotential. The electron effective mass reaches its maximum for  $T = T_C$ , and the value  $2.397m_e$  has been obtained.

The basic thermodynamic parameters characterizing the superconducting state in  $\text{Si}_2\text{H}_6$  under the pressure at 275 GPa have been determined.

It has been found that the critical temperature is very high even for the large values of the Coulomb pseudopotential:  $T_C \in (173.36, 99.99)$  K.

In next step, the low-temperature value of the energy gap at the Fermi level has been determined. On the basis of

the presented results, it has been proven that the dimensionless parameter  $R_\Delta$  greatly exceeds the value predicted by the BCS theory:  $R_\Delta \in \langle 4.40, 4.04 \rangle$ .

In the last step, the electron effective mass has been calculated. It has been shown that in the whole range of the superconducting state's existence, the value of  $m_e^*$  is high and reaches the maximum in the critical temperature:  $[m_e^*]_{T=T_C}^{\max} = 2.397m_e$ .

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